

Supporting Information

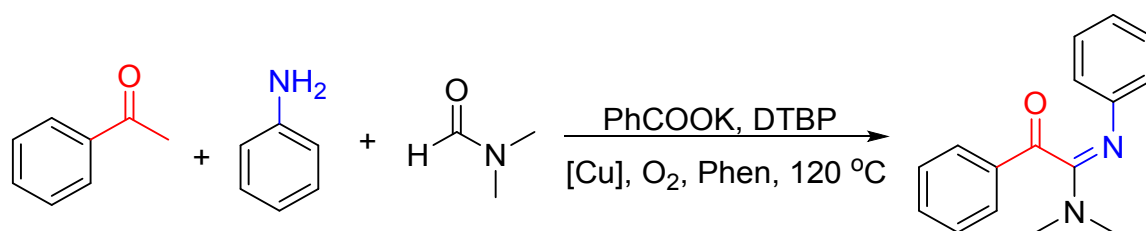
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1. General Information

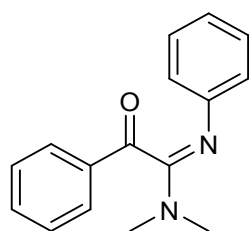
Except noted otherwise, all reactions were carried out in Schlenk tubes. Reagents and solvents were obtained from commercial sources and used without further purification. The ^1H and ^{13}C spectra were recorded on a Bruker ADVANCE III spectrometer at 400 MHz and 100 MHz, and chemical shifts were reported in parts per million (ppm). Flash column chromatography was performed using silica gel of 300-400 μm . The GC-MS results were recorded on a GC-MS QP2010 equipment, GC analysis was performed on GC 2010 Plus. The electron ionization (EI) method was used for HRMS measurement, and the mass analyzer type is TOF for EI. The HRMS (EI) was recorded on an Esquire 3000 plus instrument.

2. General Procedures

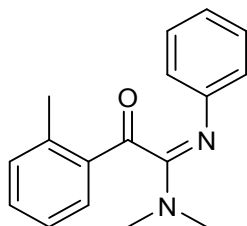


In a Schlenk tube of 25 mL, acetophenone 1 (0.5 mmol, 1.0 equiv.), aniline 2a (0.5 mmol, 1.0 equiv.), CuCl₂ (0.1 mmol, 0.2 equiv.), Phen (1,10-Phenanthroline monohydrate, 0.1 mmol, 0.2 equiv.), PhCOOK (0.5 mmol, 1.0 equiv.) and DTBP (2 mmol, 4 equiv.) were dissolved in DMF (2 mL). The mixture was stirred at 120 °C for 36 h under O₂ atmosphere. After completion of the reaction, the resulting solution was cooled to room temperature; the solution was diluted with ethyl acetate (10 mL), washed with water (5 mL), extracted with ethyl acetate (3×5 mL), and dried over anhydrous Na₂SO₄ and concentrated in vacuo. The crude product was purified by flash column chromatography on silica gel to give the desired product.

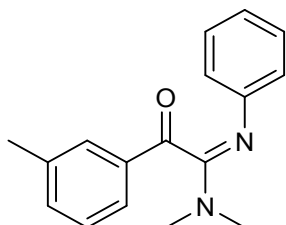
3. Spectra Data of the Products



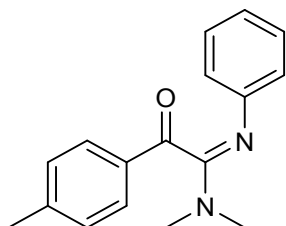
(E)-N, N-dimethyl-2-oxo-N',2-diphenylacetimidamide (4aa): Prepared according to the general procedure to afford a yellow solid in 85% yield; m. p.: 117-118 °C. ^1H NMR (400 MHz, CDCl₃) δ 7.78 (d, J = 7.6 Hz, 2H), 7.51 (t, J = 7.4 Hz, 1H), 7.38 (t, J = 7.6 Hz, 2H), 6.99 (t, J = 7.6 Hz, 2H), 6.75 (t, J = 7.4 Hz, 1H), 6.70 (d, J = 7.6 Hz, 2H), 3.03 (s, 6H). ^{13}C NMR (100 MHz, CDCl₃) δ 194.50, 156.82, 148.65, 134.48, 134.38, 129.23, 128.83, 128.30, 122.61, 122.19, 37.23. **HRMS (EI):** calcd for C₁₆H₁₆N₂O: 252.1263; found: 252.1255



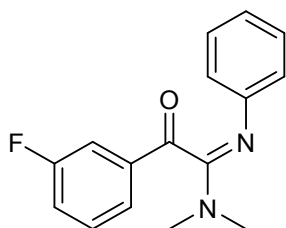
(E)-N, N-dimethyl-2-oxo-N'-phenyl-2-(o-tolyl)acetimidamide (4ba): Prepared according to the general procedure to afford a yellow solid in 80% yield; m. p.: 70-71 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.65 (d, $J = 7.6$ Hz, 1H), 7.31 (t, $J = 7.4$ Hz, 1H), 7.22 (t, $J = 7.8$ Hz, 1H), 7.06 (d, $J = 7.6$ Hz, 1H), 6.96 (t, $J = 7.4$ Hz, 2H), 6.73 (t, $J = 7.6$ Hz, 1H), 6.62 (d, $J = 7.6$ Hz, 2H), 3.06 (s, 6H), 2.32 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 196.24, 158.07, 148.89, 140.56, 133.83, 133.14, 132.11, 131.93, 128.15, 125.88, 122.65, 122.03, 37.27, 21.20. **HRMS (EI):** calcd for $\text{C}_{17}\text{H}_{18}\text{N}_2\text{O}$: 266.1419; found: 266.1413



(E)-N, N-dimethyl-2-oxo-N'-phenyl-2-(m-tolyl)acetimidamide (4ca): Prepared according to the general procedure to afford a yellow oil in 86% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.50 (s, 2H), 7.22 (d, $J = 7.2$ Hz, 1H), 7.18 (d, $J = 6.8$ Hz, 1H), 6.90 (t, $J = 7.2$ Hz, 2H), 6.64 (dd, $J = 16.4, 8.0$ Hz, 3H), 2.92 (s, 6H), 2.23 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 193.61, 155.85, 147.79, 137.65, 134.23, 133.38, 128.36, 127.67, 127.24, 125.76, 121.54, 121.07, 36.34, 20.17. **HRMS (EI):** calcd for $\text{C}_{17}\text{H}_{18}\text{N}_2\text{O}$: 266.1419; found: 266.1411

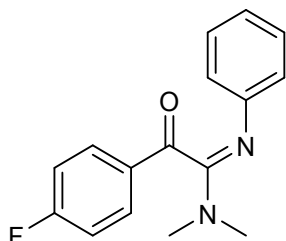


(E)-N, N-dimethyl-2-oxo-N'-phenyl-2-(p-tolyl)acetimidamide (4da): Prepared according to the general procedure to afford a yellow oil in 89% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.68 (d, $J = 7.6$ Hz, 2H), 7.18 (d, $J = 7.6$ Hz, 2H), 7.00 (t, $J = 7.4$ Hz, 2H), 6.76 (d, $J = 7.2$ Hz, 1H), 6.71 (d, $J = 7.6$ Hz, 2H), 3.01 (s, 6H), 2.35 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 194.05, 156.97, 148.83, 145.59, 132.11, 129.59, 129.42, 128.29, 122.61, 122.10, 37.23, 21.85. **HRMS (EI):** calcd for $\text{C}_{17}\text{H}_{18}\text{N}_2\text{O}$: 266.1419; found: 266.1414



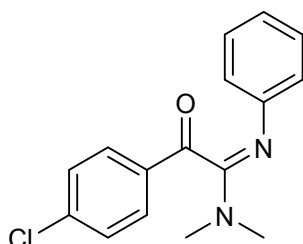
(E)-2-(3-fluorophenyl)-N, N-dimethyl-2-oxo-N'-phenylacetimidamide (4ea):

Prepared according to the general procedure to afford a yellow oil in 80% yield. ^1H NMR (400 MHz, CDCl_3) δ 7.57 (d, $J = 7.6$ Hz, 1H), 7.44 (d, $J = 8.8$ Hz, 1H), 7.36 (dd, $J = 13.8, 7.2$ Hz, 1H), 7.20 (t, $J = 8.2$ Hz, 1H), 7.00 (t, $J = 7.4$ Hz, 2H), 6.76 (t, $J = 7.2$ Hz, 1H), 6.68 (d, $J = 7.6$ Hz, 2H), 3.02 (s, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 193.44, 163.99, 161.52, 156.23, 148.46, 136.54, 136.48, 130.65, 130.58, 128.41, 125.25, 125.22, 122.54, 122.36, 121.60, 121.38, 115.5, 115.32, 37.32. HRMS (EI): calcd for $\text{C}_{16}\text{H}_{15}\text{FN}_2\text{O}$: 270.1168; found: 270.1162



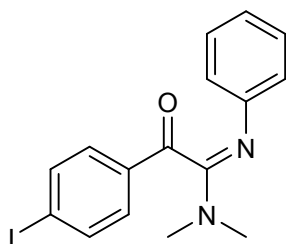
(E)-2-(4-fluorophenyl)-N, N-dimethyl-2-oxo-N'-phenylacetimidamide (4fa):

Prepared according to the general procedure to afford a yellow oil in 83% yield. ^1H NMR (400 MHz, CDCl_3) δ 7.85 – 7.73 (m, 2H), 7.06 - 6.98 (m, 4H), 6.77 (t, $J = 7.4$ Hz, 1H), 6.69 (d, $J = 7.6$ Hz, 2H), 3.03 (s, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 192.73, 167.66, 165.10, 156.54, 148.37, 132.09, 131.99, 130.96, 130.93, 128.39, 122.56, 122.38, 116.29, 116.07, 37.28. HRMS (EI): calcd for $\text{C}_{16}\text{H}_{15}\text{FN}_2\text{O}$: 270.1168; found: 270.1160



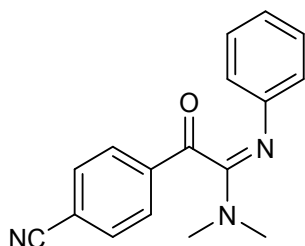
(E)-2-(4-chlorophenyl)-N, N-dimethyl-2-oxo-N'-phenylacetimidamide (4ga):

Prepared according to the general procedure to afford a yellow oil in 78% yield. ^1H NMR (400 MHz, CDCl_3) δ 7.71 (d, $J = 7.6$ Hz, 2H), 7.35 (d, $J = 7.2$ Hz, 2H), 7.01 (t, $J = 7.2$ Hz, 2H), 6.77 (t, $J = 7.2$ Hz, 1H), 6.69 (d, $J = 7.6$ Hz, 2H), 3.02 (s, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 192.22, 155.32, 147.30, 139.96, 131.77, 129.52, 128.25, 127.39, 121.52, 121.39, 36.32. HRMS (EI): calcd for $\text{C}_{16}\text{H}_{15}\text{ClN}_2\text{O}$: 286.0873; found: 286.0866



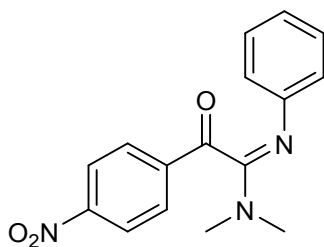
(E)-2-(4-iodophenyl)-N,N-dimethyl-2-oxo-N'-phenylacetimidamide (4ha):

Prepared according to the general procedure to afford a yellow oil in 67% yield. ^1H NMR (400 MHz, CDCl_3) δ 7.75 (d, $J = 8.0$ Hz, 2H), 7.47 (d, $J = 7.6$ Hz, 2H), 7.01 (t, $J = 7.4$ Hz, 2H), 6.79 (t, $J = 7.4$ Hz, 1H), 6.69 (d, $J = 7.6$ Hz, 2H), 3.03 (s, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 192.83, 155.23, 147.27, 137.21, 132.59, 129.32, 127.39, 121.51, 121.37, 102.09, 36.28. HRMS (EI): calcd for $\text{C}_{16}\text{H}_{15}\text{IN}_2\text{O}$: 378.0229; found: 378.0223



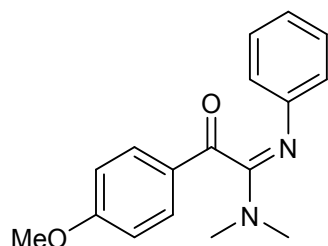
(E)-2-(4-cyanophenyl)-N,N-dimethyl-2-oxo-N'-phenylacetimidamide (4ia):

Prepared according to the general procedure to afford a yellow oil in 82% yield. ^1H NMR (400 MHz, CDCl_3) δ 7.83 (d, $J = 7.6$ Hz, 2H), 7.66 (d, $J = 7.6$ Hz, 2H), 6.99 (t, $J = 7.4$ Hz, 2H), 6.76 (t, $J = 7.4$ Hz, 1H), 6.65 (d, $J = 7.6$ Hz, 2H), 3.03 (s, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 193.40, 155.79, 147.80, 137.11, 132.66, 129.37, 128.54, 122.71, 122.55, 117.61, 117.39, 37.49. HRMS (EI): calcd for $\text{C}_{17}\text{H}_{15}\text{N}_3\text{O}$: 277.1215; found: 277.1209



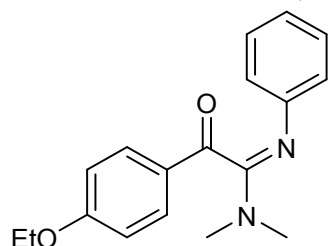
(E)-N,N-dimethyl-2-(4-nitrophenyl)-2-oxo-N'-phenylacetimidamide (4ja):

Prepared according to the general procedure to afford a yellow oil in 67% yield. ^1H NMR (400 MHz, CDCl_3) δ 8.19 (d, $J = 8.0$ Hz, 2H), 7.90 (d, $J = 8.0$ Hz, 2H), 6.98 (t, $J = 7.4$ Hz, 2H), 6.75 (t, $J = 7.2$ Hz, 1H), 6.65 (d, $J = 8.0$ Hz, 2H), 3.04 (s, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 193.33, 155.74, 150.76, 147.92, 138.51, 130.07, 128.56, 124.05, 122.68, 122.52, 37.61. HRMS (EI): calcd for $\text{C}_{16}\text{H}_{15}\text{N}_3\text{O}_3$: 297.1113; found: 297.1108



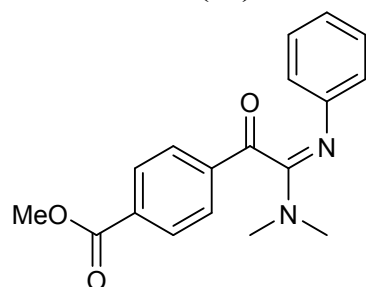
(E)-2-(4-methoxyphenyl)-N, N-dimethyl-2-oxo-N'-phenylacetimidamide (4ka):

Prepared according to the general procedure to afford a yellow solid in 89% yield; m. p.: 80-81 °C. ¹H NMR (400 MHz, CDCl₃) δ 7.76 (d, *J* = 7.6 Hz, 2H), 7.01 (t, *J* = 7.2 Hz, 2H), 6.84 (d, *J* = 7.6 Hz, 2H), 6.75 (dd, *J* = 19.2, 7.6 Hz, 3H), 3.82 (s, 3H), 3.02 (s, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 192.67, 164.53, 157.14, 148.82, 148.82, 131.79, 128.30, 127.65, 122.60, 122.13, 114.14, 55.53, 37.37. HRMS (EI): calcd for C₁₇H₁₈N₂O₂: 282.1368; found: 282.1362



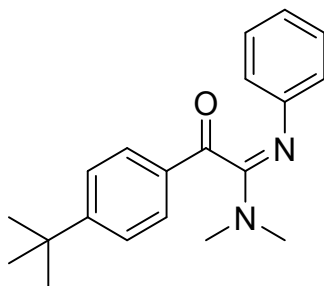
(E)-2-(4-ethoxyphenyl)-N, N-dimethyl-2-oxo-N'-phenylacetimidamide (4la):

Prepared according to the general procedure to afford a yellow solid in 81% yield; m. p.: 74-75 °C. ¹H NMR (400 MHz, CDCl₃) δ 7.68 (d, *J* = 8.0 Hz, 2H), 6.94 (t, *J* = 7.4 Hz, 2H), 6.76 (d, *J* = 8.4 Hz, 2H), 6.68 (dd, *J* = 16.6, 7.8 Hz, 3H), 3.97 (q, *J* = 6.8 Hz, 2H), 2.95 (s, 6H), 1.33 (t, *J* = 7.0 Hz, 3H). ¹³C NMR (100 MHz, CDCl₃) δ 192.70, 163.98, 157.12, 149.00, 131.78, 128.27, 127.48, 122.55, 122.03, 114.52, 63.88, 37.20, 14.61. HRMS (EI): calcd for C₁₈H₂₀N₂O₂: 296.1525; found: 296.1517



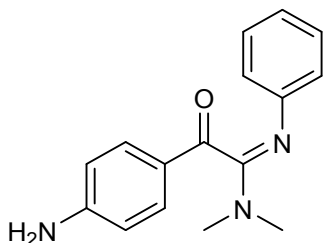
methyl (E)-4-(2-(dimethylamino)-2-(phenylimino)acetyl)benzoate (4ma):

Prepared according to the general procedure to afford a yellow oil in 69% yield. ¹H NMR (400 MHz, CDCl₃) δ 8.02 (d, *J* = 7.2 Hz, 2H), 7.81 (d, *J* = 7.6 Hz, 2H), 6.97 (t, *J* = 7.2 Hz, 2H), 6.73 (t, *J* = 7.4 Hz, 1H), 6.66 (d, *J* = 7.6 Hz, 2H), 3.90 (s, 3H), 3.03 (s, 6H). ¹³C NMR (100 MHz, CDCl₃) δ 194.21, 165.91, 156.37, 148.29, 137.39, 134.84, 130.00, 129.02, 128.41, 122.58, 122.40, 52.55, 37.40. HRMS (EI): calcd for C₁₈H₁₈N₂O₃: 310.1317; found: 310.1311



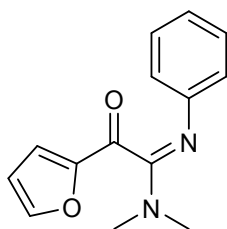
(E)-2-(4-(tert-butyl)phenyl)-N,N-dimethyl-2-oxo-N'-phenylacetimidamide (4na):

Prepared according to the general procedure to afford a yellow oil in 80% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.72 (d, $J = 7.6$ Hz, 2H), 7.39 (d, $J = 8.0$ Hz, 2H), 7.01 (t, $J = 7.2$ Hz, 2H), 6.75 (dd, $J = 16.2, 7.2$ Hz, 1H), 3.02 (s, 6H), 1.29 (s, 9H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 192.94, 157.32, 155.93, 147.67, 130.92, 128.20, 127.24, 124.81, 121.64, 121.07, 36.14, 34.24, 29.93. **HRMS (EI):** calcd for $\text{C}_{20}\text{H}_{24}\text{N}_2\text{O}$: 308.1889; found: 308.1882

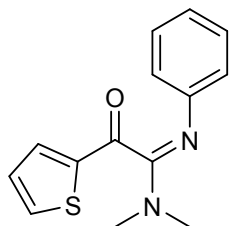


(E)-2-(4-aminophenyl)-N,N-dimethyl-2-oxo-N'-phenylacetimidamide (4oa):

Prepared according to the general procedure to afford a yellow oil in 63% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.59 (d, $J = 8.4$ Hz, 2H), 7.00 (t, $J = 7.6$ Hz, 2H), 6.77 - 6.72 (m, 3H), 6.49 (d, $J = 8.0$ Hz, 2H), 4.28 (s, 2H), 2.98 (s, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 190.79, 156.61, 151.51, 148.13, 131.10, 127.23, 123.80, 121.62, 120.97, 112.79, 36.36. **HRMS (EI):** calcd for $\text{C}_{16}\text{H}_{17}\text{N}_3\text{O}$: 267.1372; found: 267.1367

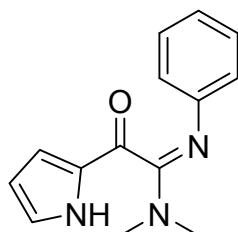


(E)-2-(furan-2-yl)-N,N-dimethyl-2-oxo-N'-phenylacetimidamide (4pa): Prepared according to the general procedure to afford a yellow oil in 92% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.52 (s, 1H), 7.07 (d, $J = 2.0$ Hz, 1H), 7.04 (t, $J = 7.4$ Hz, 2H), 6.79 (t, $J = 7.4$ Hz, 1H), 6.72 (d, $J = 7.6$ Hz, 2H), 6.42 (s, 1H), 3.01 (s, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 181.29, 155.95, 151.23, 148.71, 148.35, 128.43, 122.33, 122.30, 121.56, 112.76, 37.41. **HRMS (EI):** calcd for $\text{C}_{14}\text{H}_{14}\text{N}_2\text{O}_2$: 242.1055; found: 242.1048



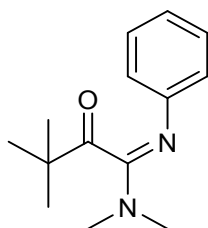
(E)-N, N-dimethyl-2-oxo-N'-phenyl-2-(thiophen-2-yl)acetimidamide (4qa):

Prepared according to the general procedure to afford a yellow solid in 88% yield; m. p.: 125-126 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.61 (t, $J = 4.0$ Hz, 2H), 7.13 – 6.98 (m, 3H), 6.79 (t, $J = 7.6$ Hz, 1H), 6.75 (d, $J = 8.0$ Hz, 2H), 3.03 (s, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 186.13, 156.47, 148.88, 142.19, 136.19, 135.80, 128.42, 122.46, 122.25, 37.38. **HRMS (EI):** calcd for $\text{C}_{14}\text{H}_{14}\text{N}_2\text{OS}$: 258.0827; found: 258.0820.

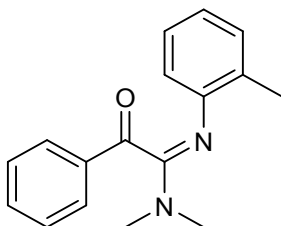


(E)-N, N-dimethyl-2-oxo-N'-phenyl-2-(1H-pyrrol-2-yl)acetimidamide (4ra):

Prepared according to the general procedure to afford a yellow oil in 45% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 9.67 (s, 1H), 7.03 (t, $J = 7.6$ Hz, 2H), 6.96 (s, 1H), 6.87 (s, 1H), 6.83 – 6.73 (m, 3H), 6.22 (q, $J = 6.2$ Hz, 1H), 3.02 (s, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 181.62, 156.93, 149.10, 130.97, 128.27, 127.03, 122.54, 122.12, 120.84, 111.65, 37.48. **HRMS (EI):** calcd for $\text{C}_{14}\text{H}_{15}\text{N}_3\text{O}$: 241.1215; found: 241.1209.

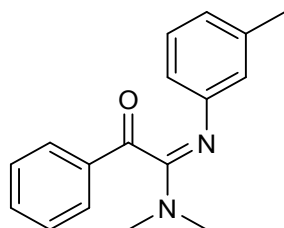


(E)-N, N,3,3-tetramethyl-2-oxo-N'-phenylbutanimidamide (4ua): Prepared according to the general procedure to afford a yellow oil in 53% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.17 (t, $J = 7.6$ Hz, 2H), 6.93 (d, $J = 7.6$ Hz, 1H), 6.78 (d, $J = 7.6$ Hz, 2H), 2.96 (s, 6H), 0.82 (s, 9H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 211.46, 156.71, 147.91, 127.56, 122.29, 121.40, 41.80, 36.35, 25.44. **HRMS (EI):** calcd for $\text{C}_{14}\text{H}_{20}\text{N}_2\text{O}$: 232.1576; found: 232.1571

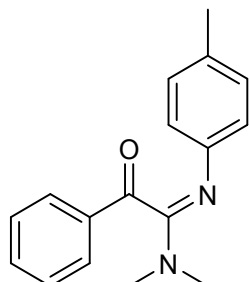


(E)-N, N-dimethyl-2-oxo-2-phenyl-N'-(o-tolyl)acetimidamide (4ab): Prepared according to the general procedure to afford a yellow solid in 77% yield; m. p.: 71-72

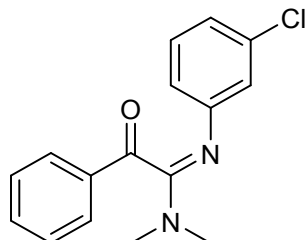
°C. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.75 (d, $J = 7.6$ Hz, 2H), 7.51 (t, $J = 7.4$ Hz, 1H), 7.38 (t, $J = 7.4$ Hz, 2H), 6.90 (d, $J = 7.2$ Hz, 1H), 6.79 (t, $J = 7.4$ Hz, 1H), 6.68 (t, $J = 7.4$ Hz, 1H), 6.51 (d, $J = 7.6$ Hz, 1H), 3.04 (s, 6H), 2.17 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 194.61, 155.97, 146.89, 134.69, 134.33, 130.32, 129.74, 128.85, 128.75, 125.65, 122.33, 121.89, 37.21, 18.37. **HRMS (EI)**: calcd for $\text{C}_{17}\text{H}_{18}\text{N}_2\text{O}$: 266.1419; found: 266.1413



(E)-N, N-dimethyl-2-oxo-2-phenyl-N'-(m-tolyl)acetimidamide (4ac): Prepared according to the general procedure to afford a yellow solid in 85% yield; m. p.: 84-85 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.78 (d, $J = 7.6$ Hz, 2H), 7.51 (t, $J = 7.2$ Hz, 1H), 7.38 (t, $J = 7.4$ Hz, 2H), 6.86 (t, $J = 7.6$ Hz, 1H), 6.58 – 6.52 (m, 2H), 6.48 (d, $J = 8.0$ Hz, 1H), 3.01 (s, 6H), 2.11 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 194.58, 156.65, 148.56, 137.87, 134.58, 134.30, 129.22, 128.78, 128.06, 123.42, 122.98, 119.51, 37.28, 21.19. **HRMS (EI)**: calcd for $\text{C}_{17}\text{H}_{18}\text{N}_2\text{O}$: 266.1419; found: 266.1411

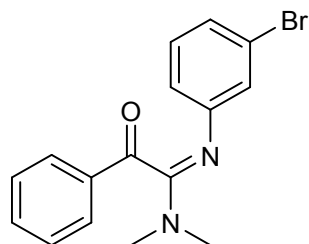


(E)-N, N-dimethyl-2-oxo-2-phenyl-N'-(p-tolyl)acetimidamide (4ad): Prepared according to the general procedure to afford a yellow solid in 86% yield; m. p.: 135-136 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.68 (d, $J = 7.6$ Hz, 2H), 7.18 (d, $J = 7.6$ Hz, 2H), 7.00 (t, $J = 7.4$ Hz, 2H), 6.76 (d, $J = 7.2$ Hz, 1H), 6.71 (d, $J = 7.6$ Hz, 2H), 3.01 (s, 6H), 2.35 (s, 3H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 194.88, 156.80, 146.02, 134.51, 134.34, 131.32, 129.25, 128.93, 128.82, 122.34, 37.33, 20.67. **HRMS (EI)**: calcd for $\text{C}_{17}\text{H}_{18}\text{N}_2\text{O}$: 266.1419; found: 266.1412



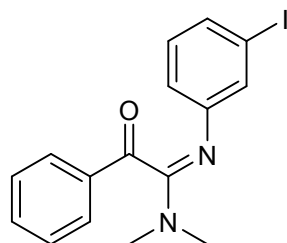
(E)-N'-(3-chlorophenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4ae): Prepared according to the general procedure to afford a yellow solid in 80% yield; m. p.: 77-78 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.76 (d, $J = 7.6$ Hz, 2H), 7.52 (t, $J = 7.2$ Hz, 1H), 7.39 (t, $J = 7.4$ Hz, 2H), 6.89 (t, $J = 8.0$ Hz, 1H), 6.71 (s, 2H), 6.56 (d, $J = 8.0$ Hz, 2H), 3.01 (d, $J = 91.2$ Hz, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 193.79, 157.02,

150.25, 134.67, 134.26, 133.66, 129.24, 129.19, 128.99, 122.84, 122.16, 120.84, 38.08, 36.48. **HRMS (EI)**: calcd for C₁₆H₁₅ClN₂O: 286.0873; found: 286.0868



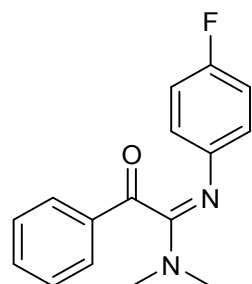
(E)-N'-(3-bromophenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4af):

Prepared according to the general procedure to afford a yellow solid in 83% yield; m. p.: 70-71 °C. **¹H NMR** (400 MHz, CDCl₃) δ 7.76 (d, *J* = 7.6 Hz, 2H), 7.52 (t, *J* = 7.4 Hz, 1H), 7.39 (t, *J* = 7.2 Hz, 2H), 6.83 (dd, *J* = 17.4, 8.8 Hz, 3H), 6.60 (d, *J* = 7.2 Hz, 1H), 3.00 (d, *J* = 87.9 Hz, 6H). **¹³C NMR** (100 MHz, CDCl₃) δ 193.75, 157.04, 150.43, 134.69, 134.27, 129.54, 129.18, 129.00, 125.75, 125.02, 121.86, 121.26, 38.04, 36.48. **HRMS (EI)**: calcd for C₁₆H₁₅BrN₂O: 330.0368; found: 330.0361



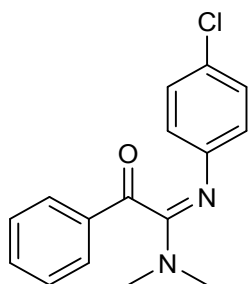
(E)-N'-(3-iodophenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4ag):

Prepared according to the general procedure to afford a yellow oil in 81% yield. **¹H NMR** (400 MHz, CDCl₃) δ 7.75 (d, *J* = 7.2 Hz, 2H), 7.52 (t, *J* = 7.4 Hz, 1H), 7.38 (t, *J* = 7.4 Hz, 2H), 7.05 (d, *J* = 13.2 Hz, 2H), 6.68 (t, *J* = 7.6 Hz, 1H), 6.63 (d, *J* = 7.6 Hz, 1H), 2.99 (d, *J* = 93.2 Hz, 6H). **¹³C NMR** (100 MHz, CDCl₃) δ 193.75 (s), 157.04 (s), 150.32 (s), 134.71 (s), 134.30 (s), 131.73 (s), 131.01 (s), 129.75 (s), 129.17 (s), 129.02 (s), 93.84 (s), 38.10 (s), 36.55 (s). **HRMS (EI)**: calcd for C₁₆H₁₅IN₂O: 378.0229; found: 378.0223



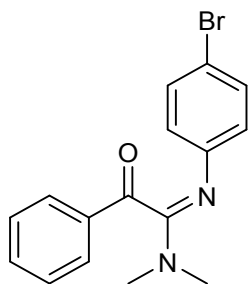
(E)-N'-(4-bromophenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4ah):

Prepared according to the general procedure to afford a yellow solid in 84% yield; m. p.: 101-102 °C. **¹H NMR** (400 MHz, CDCl₃) δ 7.75 (d, *J* = 7.6 Hz, 2H), 7.51 (t, *J* = 7.2 Hz, 1H), 7.37 (t, *J* = 7.4 Hz, 2H), 6.70 – 6.58 (m, 4H), 2.97 (s, 6H). **¹³C NMR** (100 MHz, CDCl₃) δ 194.47, 159.74, 157.35, 144.87, 144.84, 134.57, 134.32, 129.17, 128.93, 123.70, 123.62, 114.97, 114.75, 37.53. **HRMS (EI)**: calcd for C₁₆H₁₅FN₂O: 270.1168; found: 270.1163



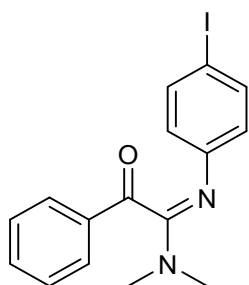
(E)-N'-(4-chlorophenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4ai):

Prepared according to the general procedure to afford a yellow oil in 87% yield. ^1H NMR (400 MHz, CDCl_3) δ 7.74 (d, $J = 7.6$ Hz, 2H), 7.49 (t, $J = 7.4$ Hz, 1H), 7.36 (t, $J = 7.4$ Hz, 2H), 6.91 (d, $J = 7.6$ Hz, 2H), 6.61 (d, $J = 7.6$ Hz, 2H), 2.98 (d, $J = 89.6$ Hz, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 194.07, 157.07, 147.51, 134.70, 134.20, 129.19, 129.01, 128.30, 127.22, 123.90, 37.95, 36.48. HRMS (EI): calcd for $\text{C}_{16}\text{H}_{15}\text{ClN}_2\text{O}$: 286.0873; found: 286.0868



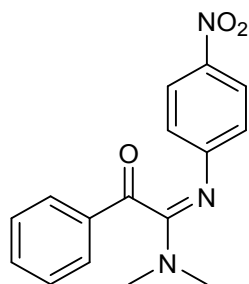
(E)-N'-(4-bromophenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4aj):

Prepared according to the general procedure to afford a yellow oil in 81% yield. ^1H NMR (400 MHz, CDCl_3) δ 7.76 (d, $J = 7.6$ Hz, 2H), 7.53 (t, $J = 7.2$ Hz, 1H), 7.40 (t, $J = 7.6$ Hz, 2H), 7.08 (d, $J = 8.0$ Hz, 2H), 6.57 (d, $J = 7.6$ Hz, 2H), 3.00 (d, $J = 79.6$ Hz, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 193.03, 155.91, 146.96, 133.65, 133.20, 130.22, 128.19, 127.97, 123.31, 113.98, 36.98, 35.54. HRMS (EI): calcd for $\text{C}_{16}\text{H}_{15}\text{BrN}_2\text{O}$: 330.0368; found: 330.0361



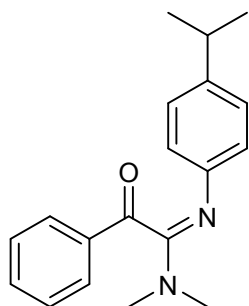
(E)-N'-(4-iodophenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4ak):

Prepared according to the general procedure to afford a yellow oil in 76% yield. ^1H NMR (400 MHz, CDCl_3) δ 7.77 (d, $J = 7.6$ Hz, 2H), 7.53 (t, $J = 7.2$ Hz, 1H), 7.39 (t, $J = 7.4$ Hz, 2H), 7.27 (d, $J = 7.6$ Hz, 2H), 6.47 (d, $J = 7.6$ Hz, 2H), 3.00 (d, $J = 94.0$ Hz, 6H). ^{13}C NMR (100 MHz, CDCl_3) δ 193.94, 156.88, 148.63, 137.21, 134.74, 134.17, 129.22, 129.04, 124.90, 85.66, 37.33, 36.48. HRMS (EI): calcd for $\text{C}_{16}\text{H}_{15}\text{IN}_2\text{O}$: 378.0229; found: 378.0224



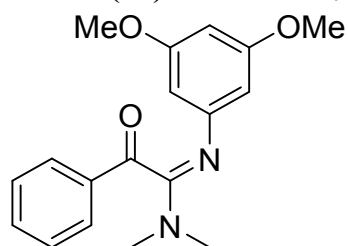
(E)-N, N-dimethyl-N'-(4-nitrophenyl)-2-oxo-2-phenylacetimidamide (4al):

Prepared according to the general procedure to afford a yellow oil in 47% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.87 (d, $J = 8.8$ Hz, 2H), 7.75 (d, $J = 7.6$ Hz, 2H), 7.53 (t, $J = 7.4$ Hz, 1H), 7.39 (t, $J = 7.8$ Hz, 2H), 6.73 (d, $J = 8.8$ Hz, 2H), 3.05 (d, $J = 102.8$ Hz, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 192.81, 156.64, 155.94, 142.47, 135.07, 133.82, 129.23, 129.19, 124.44, 122.69, 38.21, 36.53. **HRMS (EI):** calcd for $\text{C}_{16}\text{H}_{15}\text{N}_3\text{O}_3$: 297.1113; found: 297.1107



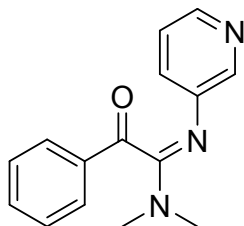
(E)-N'-(4-isopropylphenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4am):

Prepared according to the general procedure to afford a yellow oil in 86% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.76 (d, $J = 7.2$ Hz, 2H), 7.50 (t, $J = 7.4$ Hz, 1H), 7.37 (t, $J = 7.4$ Hz, 2H), 6.83 (d, $J = 6.8$ Hz, 2H), 6.61 (d, $J = 7.2$ Hz, 2H), 3.01 (s, 6H), 2.70 - 2.63 (m, 1H), 1.06 (d, $J = 6.8$ Hz, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 194.93, 156.80, 146.14, 142.51, 134.65, 134.24, 129.19, 128.75, 126.22, 122.34, 37.20, 33.25, 23.96. **HRMS (EI):** calcd for $\text{C}_{19}\text{H}_{22}\text{N}_2\text{O}$: 294.1732; found: 294.1727



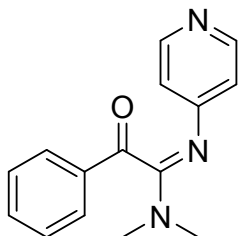
(E)-N'-(3,5-dimethoxyphenyl)-N, N-dimethyl-2-oxo-2-phenylacetimidamide (4an):

Prepared according to the general procedure to afford a yellow solid in 75% yield; m. p.: 122-123 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.79 (d, $J = 7.6$ Hz, 2H), 7.50 (t, $J = 7.4$ Hz, 1H), 7.38 (t, $J = 7.4$ Hz, 2H), 6.39 (s, 1H), 6.32 (s, 2H), 3.00 (s, 6H), 2.05 (s, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 194.62, 156.56, 148.44, 137.63, 134.71, 134.24, 129.22, 128.76, 123.97, 120.40, 37.29, 21.10. **HRMS (EI):** calcd for $\text{C}_{18}\text{H}_{20}\text{N}_2\text{O}_3$: 312.1474; found: 312.1468



(E)-N, N-dimethyl-2-oxo-2-phenyl-N'-(pyridin-3-yl)acetimidamide (4ao):

Prepared according to the general procedure to afford a yellow oil in 60% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 7.96 (s, 1H), 7.89 (d, $J = 1.2$ Hz, 1H), 7.67 (d, $J = 7.6$ Hz, 2H), 7.43 (t, $J = 7.4$ Hz, 1H), 7.30 (t, $J = 7.4$ Hz, 2H), 6.94 (d, $J = 8.0$ Hz, 1H), 6.84 (d, $J = 4.8$ Hz, 1H), 2.94 (d, $J = 115.6$ Hz, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 193.36 (s), 157.72 (s), 145.38 (s), 144.12 (s), 142.88 (s), 134.88 (s), 133.92 (s), 129.75 (s), 129.11 (s), 129.06 (s), 123.07 (s), 38.08 (s), 36.30 (s). **HRMS (EI):** calcd for $\text{C}_{15}\text{H}_{15}\text{N}_3\text{O}$: 253.1215; found: 253.1208.



(E)-N, N-dimethyl-2-oxo-2-phenyl-N'-(pyridin-4-yl)acetimidamide (4ap):

Prepared according to the general procedure to afford a yellow oil in 38% yield. $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 8.13 (d, $J = 5.6$ Hz, 2H), 7.75 (d, $J = 8.0$ Hz, 2H), 7.54 (t, $J = 7.4$ Hz, 1H), 7.40 (t, $J = 7.6$ Hz, 2H), 6.58 (d, $J = 5.6$ Hz, 2H), 3.04 (d, $J = 103.6$ Hz, 6H). $^{13}\text{C NMR}$ (100 MHz, CDCl_3) δ 192.66, 156.71, 156.52, 149.56, 134.95, 133.94, 129.24, 129.11, 118.03, 38.05, 36.37. **HRMS (EI):** calcd for $\text{C}_{15}\text{H}_{15}\text{N}_3\text{O}$: 253.1215; found: 253.1209.

4. NMR data of products

